Tracking the Isotopes

by Jere D. Knight and James E. Sattizahn

T he story of nuclear and radiochemistry at Los Alamos is a story that reflects in many ways the growth and development of the Laboratory itself. It began in the early days before the test of the first nuclear explosive, when the Laboratory's overriding concern was to design and build such an object and to ascertain how well it would work, if at all. A group of radiochemists was set up under R. W. Dodson, and as soon as they could get their laboratories and equipment in running order, there was an urgent need for their services on tasks such as the preparation and calibration of radioactive sources and the measurement of fission products, as well as for their ideas on the application of radiochemistry to other problems. Later, as design ideas began to take shape in the form of full-scale models of the explosive system, large-scale radiochemical preparations were required, preparations far outside the scale usually dealt with in university laboratories. A well-known example was the chemical processing and mounting of lanthanum-140 sources emitting trillions of gamma rays per second. These sources were used for short-time-scale radiography of imploding systems, the so-called RaLa (for radioactive lanthanum), or Bayo Canyon, experiments.

Implosion was one of the ways chosen to achieve criticality in a nuclear bomb: a ball of fissionable fuel, say plutonium, was to be squeezed into a critical configuration by detonating a carefully tailored surrounding layer of conventional high explosive. The unexploded system resembled a very large apricot in which the plutonium and an intermediate layer of dense, neutron-reflecting metal are the pit. An important question was how rapidly, how uniformly, and how deeply the pit was squeezed when the high explosive was set off. A relatively simple way to study this process was to employ radiography on a similar configuration in which the plutonium was replaced by a nonfissionable but comparably dense metal. If one could somehow have a very strong gamma-ray source at the center of the pit and an array of gamma-ray detectors outside the complete bomb, one could measure the flux of gamma rays reaching the outside as a function of time during the course of the

			No 25 0.25 ms sf
Md 247 2.9 s α	Md 248 7 s ec, α	Md 24 24 s ec, α	
Fm 246 1.1 s α, sf	Fm 247 9.2 s α | 35 s α, ec	Fm 24 36 s α, sf	
Es 245 1.3 m ec, α	Es 246 7.7 m ec, α	Es 247 4.7 m ec, α	
Cf 244 19.7 m α	Cf 245 43.6 m ec, α	Cf 246 35.7 h α, sf	
Bk 243 4.5 h ec, α	Bk 244 4.35 h ec, α	Bk 24 4.90 d ec, α	
Cm 242 162.8 d α, sf	Cm 243 28.5 y α, ec	Cm 24 18.11 y α, sf	
Am 241 432.6 y α, sf	Am 242 141 y IT | 16 h β−, ec	Am 24 7370 y α, sf	
Pu 240 6550 y α, sf	Pu 241 14.4 y β−, α	Pu 24 3.763 × α, sf	
Np 239 2.355 d β−	Np 240 7.22 m β− | 65 m β−	Np 24 16.0 m β−	
U 238 4.468 × 10⁹ y α	U 239 23.5 m β−	U 240 14.1 h β−	
Pa 237 8.7 m β−	Pa 238 2.3 m β−		
Th 236 37.1 m			

Nuclide	Half-life & decay
Lr 253	~2.6 s; α
Lr 254	16 s; α
Lr 255	21.5 s; α
Lr 256	25.9 s; α
Lr 257	0.65 s; α
Lr 258	4.35 s; α
Lr 259	5.4 s; α
Lr 260	3 m; α

Nuclide	Half-life & decay
(…) 251	s
No 252	2.3 s; α, sf
No 253	1.7 m; α
No 254	0.28 s IT | 55 s α, sf
No 255	3.1 m; α, ec
No 256	3.1 s; α, sf
No 257	26 s; α
No 258?	1.2 ms; sf
No 259	58 m; α, ec

Nuclide	Half-life & decay
(…) 250	s; α
Md 251	4.0 m; ec, α
Md 252	2.3 m; ec
Md 254	10 m ec | 28 m ec
Md 255	27 m; ec, α
Md 256	1.3 h; ec, α
Md 257	5.0 h; ec, α
Md 258	43 m ec | 56 d α
Md 259	95 m; sf

Nuclide	Half-life & decay
(…) 249	m; α
Fm 250	1.8 s IT | 30 m α, sf
Fm 251	5.30 h; ec, α
Fm 252	25.4 h; α, sf
Fm 253	3.0 d; ec, α
Fm 254	3.24 h; α, sf
Fm 255	20.1 h; α, sf
Fm 256	2.63 h; sf, α
Fm 257	100.5 d; α, sf
Fm 258	0.38 ms; sf
Fm 259	1.5 s; sf

Nuclide	Half-life & decay
(…) 248	m; α
Es 249	1.70 h; ec, α
Es 250	2.22 h ec | 8.6 h ec
Es 251	33 h; ec, α
Es 252	471.7 d; α, ec
Es 253	20.4 d; α, sf
Es 254	39.3h β⁻, α, ec | 275.7d α
Es 255	39.8 d; β⁻, α, sf
Es 256	7.6 h β⁻ | 22 m β⁻

Nuclide	Half-life & decay
(…) 247	1 h; α
Cf 248	333.5 d; α, sf
Cf 249	350.6 y; α, sf
Cf 250	13.08 y; α, sf
Cf 251	898 y; α
Cf 252	2.64 y; α, sf
Cf 253	17.81 d; β⁻, α
Cf 254	60.5 d; sf, α
Cf 255	~1 h?; β⁻
Cf 256	12.3 m; sf

Nuclide	Half-life & decay
(…) 246	0 d
Bk 247	1380 y; α
Bk 248	>9 y | 23.7 h β⁻, ec
Bk 249	320 d; β⁻, α, sf
Bk 250	3.217 h; β⁻
Bk 251	56 m; β⁻

Nuclide	Half-life & decay
(…) 245	00 y
Cm 246	4730 y; α, sf
Cm 247	1.56×10^7 y; α
Cm 248	3.397×10^5 y; α, sf
Cm 249	64.2 m; β⁻
Cm 250	$\leq 1.13 \times 10^4$ y; sf
Cm 251	16.8 m; β⁻

Nuclide	Half-life & decay
(…) 244	m | 10.1 h β⁻
Am 245	2.05 h; β⁻
Am 246	25 m β⁻ | 39 m β⁻
Am 247	22 m; β⁻

Nuclide	Half-life & decay
(…) 243	56 h
Pu 244	8.26×10^7 y; α, sf
Pu 245	10.5 h; β⁻
Pu 246	10.85 d; β⁻

Nuclide	Half-life & decay
(…) 242	m | 5.5 m β⁻

Nuclide	Half-life & decay
U 242	16.8 m; β⁻

Data from the fifth edition of Karlsruher Nuklidkarte.

Since the earliest days the Laboratory's nuclear and radiochemists have been tracking isotopes to assess the performance of nuclear weapons. Today their interests also include geochemistry, biomedicine, and basic nuclear science.

explosion. These data would provide information on the change in amount of dense absorber between the source and the detectors and thus on the change in configuration of the pit caused by the implosion. The key to this diagnostic scheme was to find such a gamma-ray source. The fission products barium-140 and lanthanum-140 (the same isotopes that led to the discovery of fission) provided the solution. Barium-140 has a half-life of 12.8 days and decays to lanthanum-140, which has a 40-hour half-life and emits an energetic gamma ray with its decay. As the operation was finally worked out, the lanthanum-140 was extracted from a large batch of its barium-140 "parent" (obtained from Oak Ridge) and put into a compact receiver in the center of the pit to be tested. Chemical separation and further purification of the lanthanum-140 were carried out by a group headed by Gerhart Friedlander at a temporary building a mile or two from the firing site in Bayo Canyon. There were then no elegant "hot cells" nor sophisticated remote-handling apparatus. To minimize radiation exposure, the radiochemists rigged up the chemical processing equipment so that the essential operations could be performed and monitored from a distance with a system of cables, mirrors, and telescopes. The final lanthanum-140, concentrated into a volume of 0.1 milliliter, was then heavily shielded and trucked to the firing site, where another simple remote-control rig transferred it into the implosion test assembly. The source, implosion assembly, and gamma-ray detectors were destroyed in the test explosion, but the barium-140 supply back at the processing site was available to serve another day: because of its relatively long half-life it could "grow in" one or more new lanthanum-140 sources for use a few days later. The RaLa implosion tests became a regular diagnostic practice; with steady improvements in technique and with increasingly strong sources, they were continued until 1962, when they

What Are Radiochemistry and Nuclear Chemistry?

T he definition of the terms "radiochemistry" and "nuclear chemistry," and the delineation of the presumed difference between them, is a problem that bedevils the toilers in this vineyard whenever they try to tell what they do. Historically, radiochemistry appears to have been the first in line. When Madame Curie was carrying through her laborious chemical procedures to isolate and identify the radioactive elements and to establish their transformations, she was doing radiochemistry. It was not until some time later that the nature of the nucleus and its reactions became clear. In most of the early researches, chemical manipulations were essential, and by the late 1930s a recognizable body of techniques and strategies had evolved to deal with the rapidly growing list of radioactive species. And it was by such chemical manipulations that Otto Hahn and Fritz Strassmann first demonstrated the occurrence of nuclear fission. Examining the products resulting from neutron irradiation of uranium, with an atomic weight around 238, they found unmistakable evidence for radioactive barium with an atomic weight around 140; they were forced to conclude that the barium came from the uranium by a process that could only have been some kind of a splitting of the uranium nucleus. Their employment of chemical techniques to study nuclear phenomena may be considered nuclear chemistry.

Unfortunately for those who would compartmentalize science into neat bins, our subject has broadened greatly in the span of time since the discovery of nuclear fission. Nowadays, its practitioners range from the "pure" radiochemists, whose primary concern is with the chemistry involved, to some "nuclear chemists" who are concerned only with nuclear physics problems and who rarely get their hands on any radioactivity—or vice versa. In our article we occupy something of a middle ground. Those pursuits in which the chemical operations are clearly of first importance we call radiochemistry, and those in which the problems of nuclear structure and reactions are of first importance we call nuclear chemistry. If in telling our story we turn out to have been inconsistent in employing this criterion, we plead guilty and ask our radiochemist/nuclear chemist critic to cast the first stone. ■

Wartime photo showing transfer of a heavily shielded gamma-ray source at the RaLa chemical processing site in Bayo Canyon. Motive power is being supplied by group leader Gerhart Friedlander and anchorperson Norma Gross, a WAC assigned to Los Alamos. Such sources were used to study the process of implosion, a method for achieving a critical configuration of the fissionable fuel in a nuclear weapon.

were supplanted by other diagnostic schemes.

The Trinity Test

The task that was to become the dominant occupation and responsibility of the nuclear and radiochemists up to the present day was one that began with the first nuclear explosion, the Trinity test of July 16, 1945 at the Jornada del Muerto site in central New Mexico. That task was the measurement of the efficiency of the explosion.

The efficiency of a nuclear explosion, like that of an industrial process such as the conversion of coal into electrical energy, is measured by the fraction of the energy content of the valuable starting material, the fuel, that appears as usable output energy. Thus, in the case of an explosion produced by nuclear fission of a fuel such as plutonium-239, the efficiency is given by the fraction of the plutonium-239 originally in the device that actually underwent fission, or "burned." The nuclear chemist proceeds to derive this fraction by gathering a small

sample of the debris from the explosion and performing radiochemical separations to isolate, individually, the plutonium and at least one radioactive fission product. By measuring the plutonium sample in an alpha-particle counter, he obtains the number of remaining plutonium nuclei—those that did not fission—which we shall call p. By measuring the fission-product sample in a beta- or gamma-ray counter, he obtains the number of fission-product nuclei. He then employs an empirical constant to convert this number to the number of fissions that had occurred in the debris sample, a quantity we shall designate by f. (The empirical constant is derived by measuring the same fission product in a calibration standard of plutonium-239 in which a known number of fissions has been induced.) The efficiency of the explosion, ϵ, is then given by

$$\epsilon = \frac{\text{fissions}}{\text{original } ^{239}\text{Pu nuclei}}$$

$$= \frac{\text{fissions}}{\text{fissions} + \text{remaining Pu nuclei}}$$

$$= \frac{f}{f + p} .$$

Thus, we see that we are able to determine the explosion efficiency by measuring the fission products and the unburned fuel in a small fraction of the explosion debris *without having to know explicitly the fraction of the total debris represented in that sample.* In actual practice we can perform this measurement on only ten trillionths of the total debris created by a 10-kiloton test, if the efficiency is the only performance parameter desired. Also, in actual practice there are a number of nontrivial complications that we have swept under the rug: some of the plutonium-239 is consumed by the nonfission reaction ^{239}Pu $(n,\gamma)^{240}\text{Pu}$ (that is, plutonium-239 + neutron → gamma rays + plutonium-240); some additional plutonium-239 may have been

created by neutron reactions with any uranium-238 present in the device followed by beta decay [$^{238}U(n,\gamma)^{239}U \xrightarrow{\beta^-} {}^{239}Np \xrightarrow{\beta^-} {}^{239}Pu$]; and chemical fractionation may have caused our sample to become enriched or depleted in the fission product chosen for analysis, so that f and p may not be found in the ratio in which they originally occurred. There are methods for dealing with these and other complications.

By multiplying the efficiency by the number of plutonium-239 nuclei known to have been put in the device, we can calculate the total number of fissions that occurred in the explosion. This is the number that the nuclear chemist actually determines, and it can easily be converted into a yield, or total energy released. For practical reasons nuclear explosion yields have traditionally been expressed in terms of tons or kilotons of TNT equivalent. Conversion of the chemist's number to kilotons involves multiplying total fissions by a nominal energy release per fission and inclusion of minor additional energy contributions from sources such as radiative neutron capture. There are other measures of the energy release in a test explosion (fireball expansion rate, atmospheric pressure wave, underground seismic signal, and so on) but the direct nuclear chemistry measurements provide the standard energy release to which all the other measurement techniques are calibrated.

The determination of the energy released in the Trinity explosion employed just these principles and measurements, albeit with equipment that would be considered primitive by modern standards. The job was performed by a team headed by Herbert Anderson and Nathan Sugarman, the latter on temporary assignment from the Manhattan Project at the University of Chicago. Their final report of the measurement, dated September 25, 1945 and still bearing a secret classification, contains some interesting notes on the planning and execution of this unprecedented operation. Since it was recognized that, if the explosion went as it was
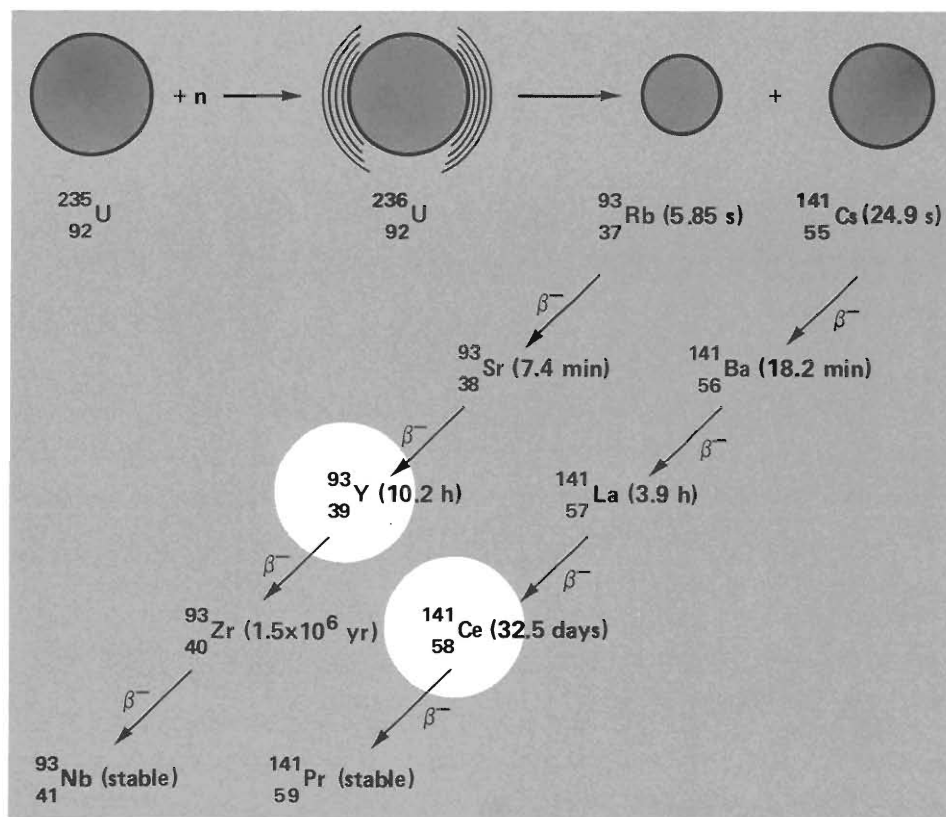
The energy generated by both nuclear power reactors and so-called atomic bombs arises from the neutron-induced fission of certain isotopes such as uranium-235 and plutonium-239. Shown here is a typical fission event: absorption of a neutron by uranium-235 leads to an excited state of uranium-236, which splits into two primary fission products, in this event rubidium-93 and cesium-141, within about 10^{-21} second. (Not shown are the additional neutrons emitted as the excited nucleus splits; these neutrons make the fission reaction a self-sustaining, or chain, reaction.) Each of the neutron-rich primary fission products undergoes a series of beta decays leading eventually to a stable isotope. As the residues of a nuclear weapon test, the fission products contain information about the weapon's performance. For example, the number of fissions that occurred can be determined by assaying a sample of the explosion debris for one or more of the fission products. A radiochemist setting out to do this by assaying for one of the fission products in the decay chains shown here would most probably pick yttrium-93 or cerium-141. The preceding chain members decay too rapidly, and the subsequent members decay too slowly (or not at all). The excited uranium-236 nucleus can also split initially into any one of a large set of possible primary fission-product pairs. The resulting mixture of primary fission products and their decay products, which contains various isotopes of about three dozen elements, is the almost universal diet of Los Alamos radiochemists. Although a nuisance when a particular isotope must be isolated, the fission-product mixture is a rich lode of material for basic research in nuclear chemistry, including investigations of the fission process and studies of individual nuclides on the neutron-rich side of stability. Such studies at Los Alamos have resulted in the discovery of at least fifteen new nuclides with half-lives ranging from less than a second to millions of years.
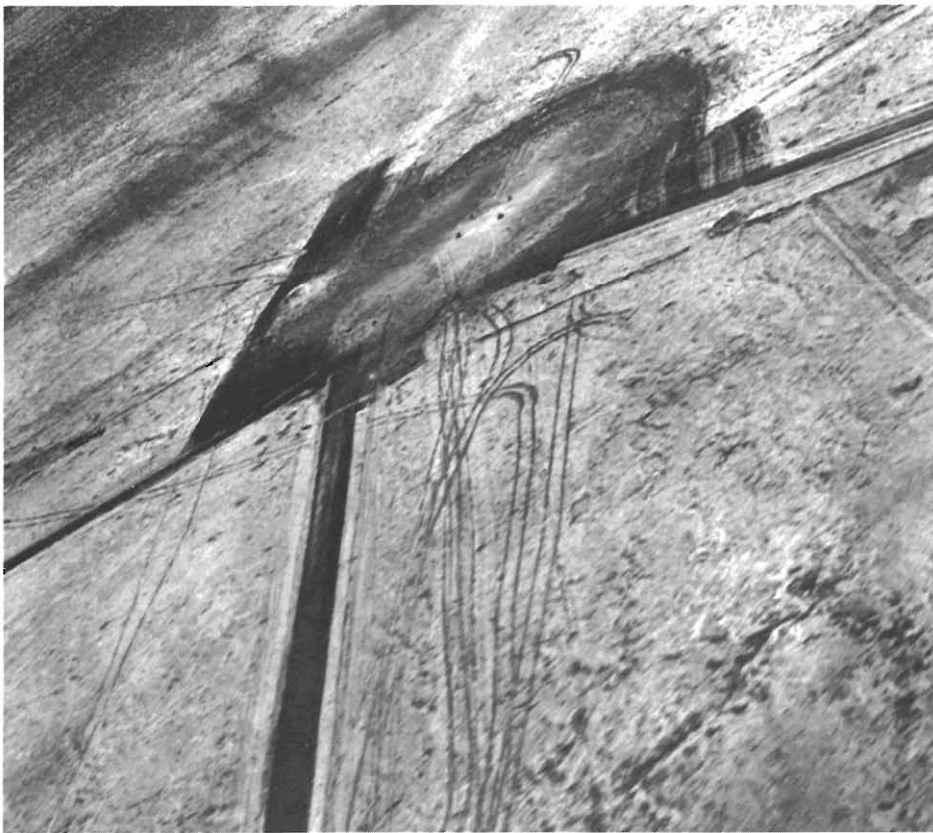
The crew that collected samples of radioactive debris from the Trinity crater rode to its destination in a Sherman tank outfitted with lead shielding and bottles of breathing air. The samples were gathered through a trapdoor in the bottom of the tank. The lower photo is an aerial view of the crater and the tracks left by the tank.

supposed to, the best samples would probably be found in the ground beneath the blast, means had to be devised to get at this material. Two Sherman tanks were made available for the purpose. One of them was fitted with lead-shielded compartments for the driver and the observer/sampler, who gathered earth samples through a hole in the floor of his compartment with a vacuum cleaner or by driving a hollow pipe into the ground. The second tank was equipped to fire rockets into the center of the crater from a distance of 500 yards; the rockets were fitted with sampling noses and with cables for retrieval. Both methods worked, and a good set of samples was brought back to Los Alamos for processing and radiochemical analysis.

One's first reaction on reading these accounts today is astonishment that such measurements could be made with useful accuracy at all, given the primitive instruments available at the time. Apparatus now considered indispensable—high-resolution gamma-ray detectors, multichannel pulse-height analyzers, and stable transistorized electronics, to name just a few—did not exist. In point of fact, the efficiency and yield numbers generated by this work were probably accurate to ± 10 percent, a quite respectable error even today. The nontrivial lesson to be learned is that ingenuity and scientific craftsmanship can achieve impressive results even with rudimentary equipment.

More Weapons Testing and a Radiochemistry Group

The experience gained in the Trinity test was soon put to use in the summer of 1946, when similar diagnostic measurements were made on the two test explosions of Operation Crossroads at Bikini Atoll in the Marshall Islands. There was still no specifically designated nuclear and radiochemistry group at Los Alamos, most of the experienced personnel having left in the great diaspora following

the end of World War II. Thus, when the call came for a working group to carry out the task at Crossroads, a team of experienced individuals was gathered from Los Alamos, Argonne, Oak Ridge, and various universities under the leadership of William "Buck" Rubinson.

This time, two kinds of postexplosion debris samples were gathered: from the atmosphere, by drone B-17 and F-6F aircraft, and from the sea, by drone boats. To provide early information in the field, a temporary radiochemical laboratory with appropriate radiation counting equipment was set up nearby at Kwajalein Atoll. The shorter lived products from the explosion (such as zirconium-97 and molybdenum-99 from fission and neptunium-239 from neutron capture on uranium-238) were analyzed at Kwajalein, and the longer lived products (strontium-89, zirconium-95, barium-140, and cerium-144 from fission) were analyzed in samples taken back to Los Alamos. The total plutonium content of the individual samples was measured at both laboratories, but measuring the ratio of plutonium-239 to total plutonium, which involved fission counting at a reactor, was of course done at Los Alamos. [This ratio provides information about the fraction of a weapon's original plutonium-239 content that is "wasted" in nonfission neutron reactions such as $(n,2n)$ and (n,γ).]

The fission-product measurements for Operation Crossroads showed for the first time in a practical way the consequences of the occurrence of noble-gas elements in fission-product decay chains. Fission products such as strontium-89 and barium-140, which derive in large part from the noble-gas ancestors 3.15-minute krypton-89 and 13.7-second xenon-140, did not "track" well with other fission products derived from chains with very short-lived or insignificant noble-gas ancestry. It was clear that even less than a minute spent as a gas during decay of a predecessor was enough to cause significant chemical fractionation of

Samples of airborne debris from the Operation Crossroads tests of 1946 were gathered by drone aircraft. Shown here is a sampling unit being installed behind the cockpit of a drone B-17. The filter paper in the sampling unit was specially treated so that the debris would stick to its surface.

that fission product relative to others and to neptunium and plutonium. No way has ever been found to exploit this fact for study of bomb-cloud phenomenology; its only effect has been to restrict the choice of fission-product nuclides suitable for weapons test diagnostics.

The period beginning in late 1946 and early 1947 saw the formation at Los Alamos of a permanent nuclear and radiochemistry group led by R. W. Spence and soon to become a part of J Division, the test organization. This change had two important effects. First, it provided the organizational focal point for dealing in a more coherent way with the spectrum of contributions that nuclear and radiochemists could make to a weapons development laboratory. More important from a scientific standpoint, it set the stage for the transition of the nuclear chemists from their status as primarily consumers of basic science information to producers as well.

Operation Sandstone in 1948, in which three new experimental fission-weapon designs were tested at Eniwetak Atoll, was an important landmark for the new group and for the entire Laboratory. The successful conduct of these tests and the information gained from them confirmed to the defense

establishment and to the fledgling Atomic Energy Commission the importance of continuing Los Alamos as an integral part of the national defense research and development effort. From a more technical standpoint these tests showed that radiochemical analyses of atmospheric samples alone could provide essential efficiency information and in addition detailed information on the performance of individual components of the test device. The demonstrated success of air sampling followed by radiochemical analysis was important not only to the nation's domestic nuclear weapons testing program but also to the development of its atmospheric surveillance system for detecting and evaluating possible foreign weapons tests. It was such a system that picked up evidence of a nuclear event in the Soviet Union in August 1949, and radiochemical analysis of the evidence was able to show that the event was indeed the test of a nuclear explosive device.

Continued Growth and the Mike Test

Following the Operation Sandstone tests, and with them the assurance of a continuing contribution to the Laboratory's future, members of the newly organized nuclear and
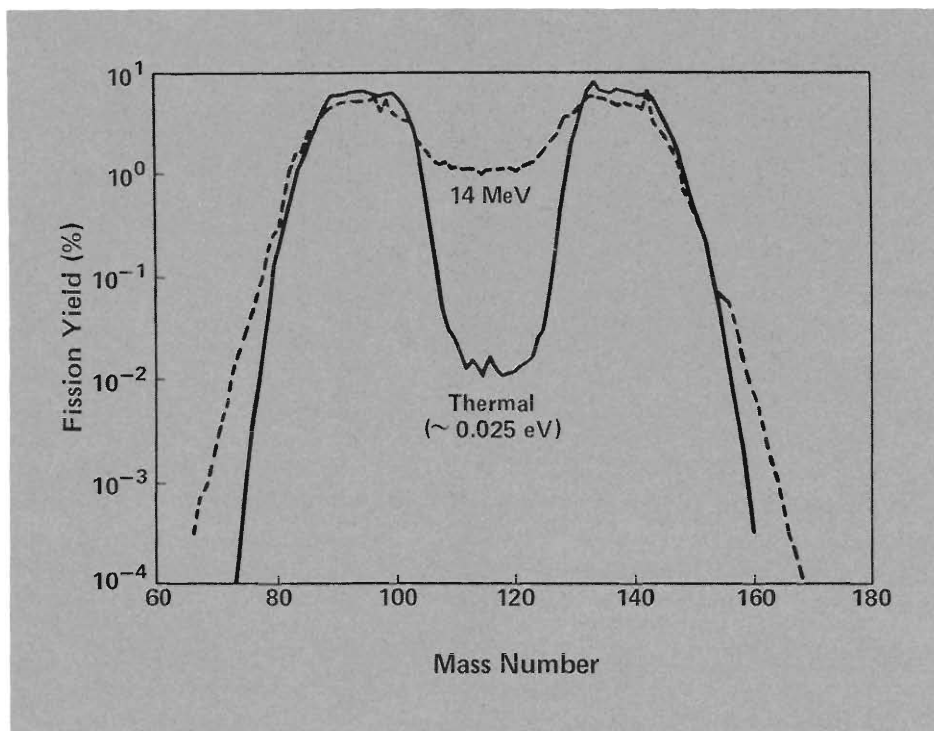
The neutron-induced fission of uranium-235 results in a wide range of products. The graph above shows that the yield of these fission products has a relative minimum in the mass region corresponding to symmetric and nearly symmetric fission and that the yields in this region increase with the energy of the neutrons inducing the fission. Yield curves for other fissile isotopes exhibit similar shapes and variations with neutron energy.

radiochemistry group (now a part of J Division) began a program of research and development along two complementary lines. First and perhaps the most immediately urgent was development of radiochemical procedures, instrumentation, and calibrations to support both programmatic functions and research in this field. The second was the research itself. Of particular interest was the study of fission-product yields and decay properties. Careful measurements were made of the mass distributions of fission-product yields from the fission of uranium-235, uranium-238, and plutonium-239 induced by various reactor neutron spectra. Similar measurements were made for fission induced by monoenergetic 14-MeV neutrons. [Neutrons with this energy are a product of the reaction $^3H(^2H,n)^4He$ induced by bombarding targets containing tritium (hydrogen-3) with deuterons (hydrogen-2 ions) from the Laboratory's Cockcroft-Walton accelerator. The same reaction between tritium and deuterium at thermonuclear temperatures is the dominant reaction and energy source for controlled thermonuclear power.] The latter results, subsequently reported at one of the Gordon Research Conferences on Nuclear

Chemistry, are believed to be the first of their kind. Studies were also begun on the decay properties of individual fission products and a variety of other nuclides and on nuclear cross sections, in particular cross sections for reactions with 14-MeV neutrons. Some of this work led in 1949 to the group's first discovery of a new radionuclide—an isomeric state of niobium-97 denoted by ^{97m}Nb. The scheme for decay of zirconium-97 to ^{97m}Nb and its decay to the ground state was reported in *Physical Review* and became the first in a large number of research papers to be published by Los Alamos nuclear chemists and their collaborators from other groups and institutions.

An important part of the Laboratory's scientific history, and one that contributed significantly to the development of nuclear and radiochemistry, was the creation in 1949 of a special arrangement with the University of New Mexico to provide scientific instruction at Los Alamos. As part of this program, Professor Milton Kahn of the University's Chemistry Department began commuting from Albuquerque to give a two-semester night-school course entitled "Chem. 213. Radiochemistry." This program helped fill an urgent need: an opportunity for Labora-

tory employees to carry out graduate-level studies and eventually to earn advanced degrees while continuing their active commitment to ongoing Laboratory programs. Though the Ph.D. requirements included an academic year in residence at Albuquerque, in practice a candidate could and did return frequently to Los Alamos to take part in the activities of his group. After the year of residence, the candidate returned to Los Alamos to do his thesis research; the topic was chosen to be relevant to Laboratory interests, and the progress of the work was guided jointly by Professor Kahn and a Laboratory staff member. A total of thirteen University of New Mexico doctorates were awarded on radiochemistry and nuclear chemistry theses under this program. Doctoral thesis work of students from the Massachusetts Institute of Technology, the University of Wyoming, and the University of California, San Diego has also been performed here under similar arrangements.

The major expansion in the Laboratory's weapons design and testing effort that began in 1950 had a corresponding impact on the size and scope of the group's activity. The first Nevada test series in early 1951, a test series at Eniwetak in the spring of 1951, and another Nevada test series in the fall of that same year presented the group with the sharp challenge not only of providing diagnostic information on a large number of fission explosions, but also of exploring and eventually developing means for obtaining quantitative diagnostic information on the phenomena associated with thermonuclear fusion components. The latter led to the first use of "radiochemical detectors": small amounts of suitable target elements incorporated into or near the device components to provide, by their level of activation, a measure of the neutron spectrum and fluence at these sites. The success of these early detector trials has led in succeeding years to a steady growth in the scale and sophistication of this diagnostic tool, to the point where nowadays the detector informa-

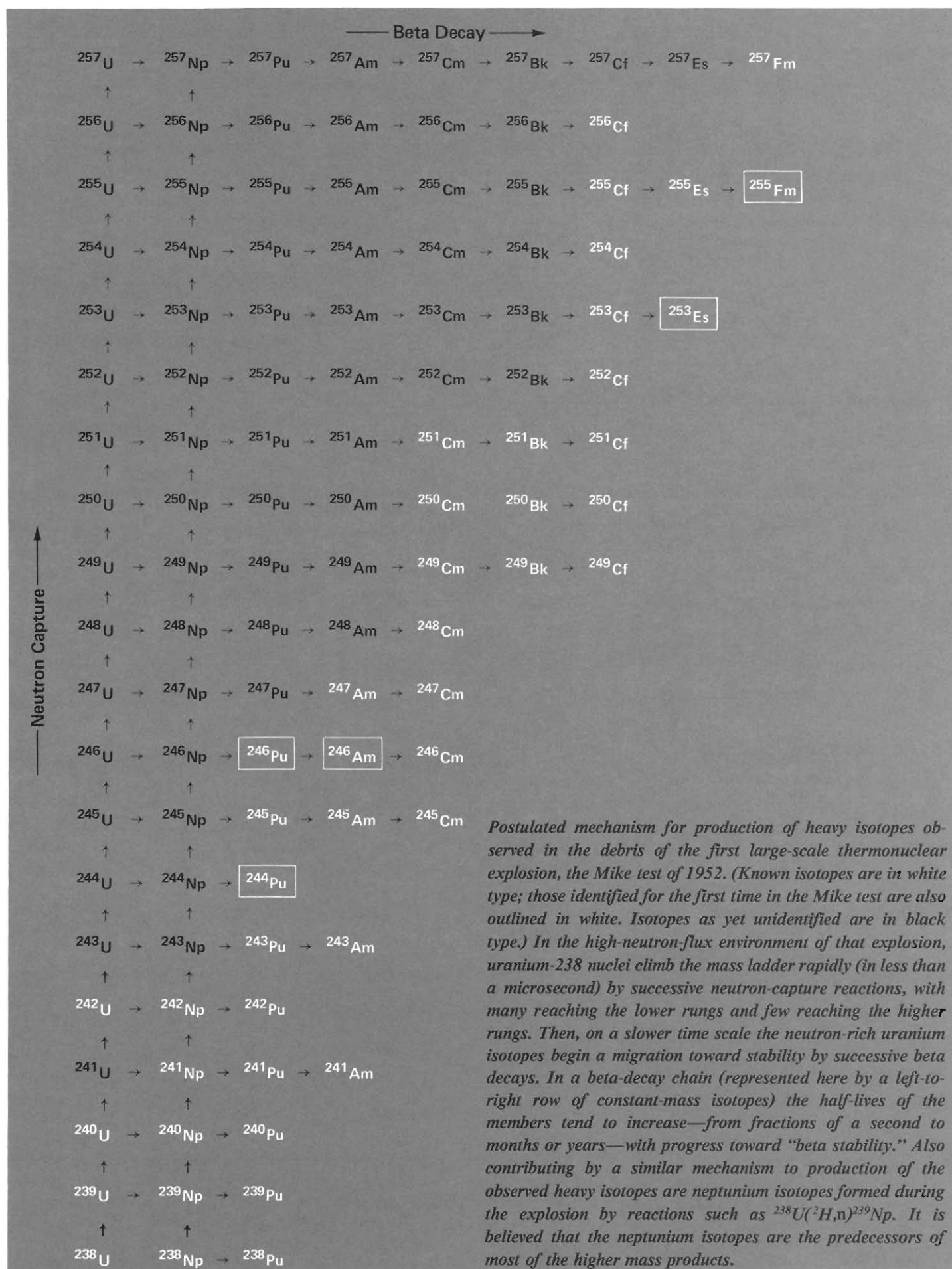tion is the most important radiochemical contribution to weapons testing.

The first large-scale thermonuclear explosion, the Mike test of 1952, handed the nuclear chemists an unexpected bonus: from its unprecedented neutron fluxes came a grand assortment of transuranium nuclides, including two new elements. Using samples of airborne debris collected by aircraft from the Mike cloud, a collaboration of nuclear chemists from the University of California Radiation Laboratory, Argonne National Laboratory, and Los Alamos undertook a thorough analysis of the transuranium elements by means of cation-exchange resin separations. These experiments showed a number of previously unknown radioactive species, among them an isotope of element 99 emitting 6.6-MeV alpha particles and a 7.1-MeV alpha emitter growing from an element-99 parent. Further work identified the first of these activities as $^{253}99$ produced by beta decay of californium-253 and the second as $^{255}100$ arising from beta decay of $^{255}99$. The new elements 99 and 100 were eventually named einsteinium and fermium, respectively. Also discovered in the Mike debris were the new nuclides plutonium-244 (the longest lived of the plutonium isotopes) and the beta emitters plutonium-246 and americium-246. Detailed examination of the abundances of the transuranium isotopes revealed what had happened. Some of the uranium in the device, together with some of the neptunium produced by various nuclear reactions during the explosion, was exposed to neutron fluxes high enough to produce strings of successive neutron capture reactions leading to uranium and neptunium isotopes with masses sixteen units or more heavier than those of the starting isotopes; these neutron-rich product isotopes then underwent successive beta decays culminating in the nuclides later observed by the radiochemists. Indeed, because of the new family of nuclear phenomena made possible by the great neutron densities achieved in the thermonuclear burn, the Mike debris turned
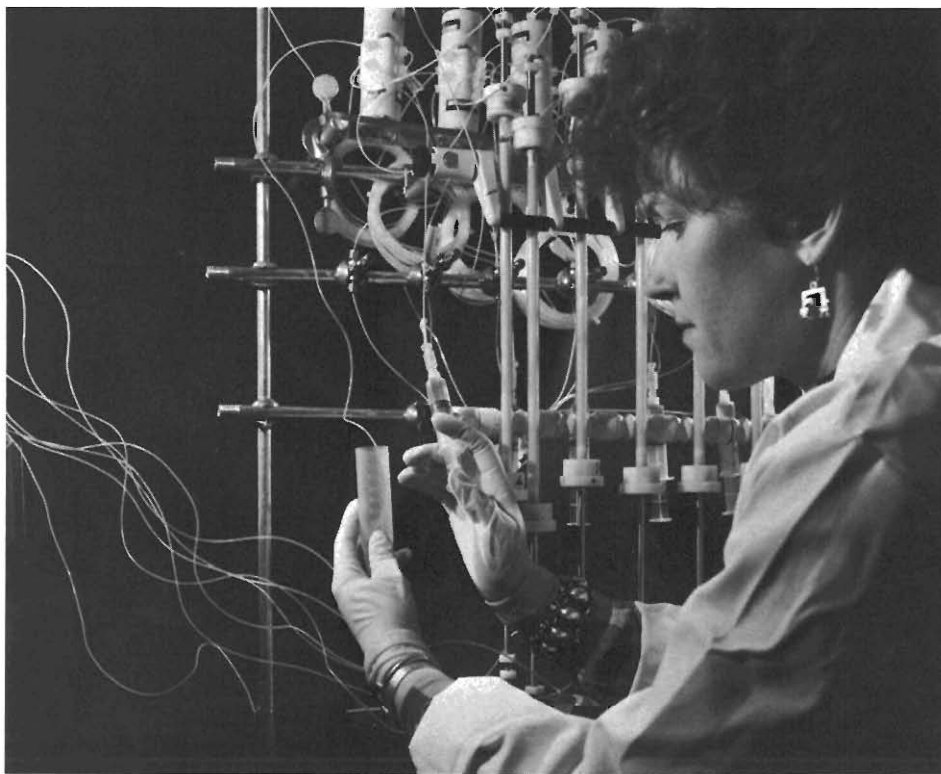
Ion exchange is a powerful technique for separating chemically similar elements. Chemist Deva Handel here loads an ion-exchange column with a solution of mixed rare earths. The columns (light vertical tubes at right center) contain a resin similar to the zeolite in water softeners. Flow of a complexing agent causes the rare earths to migrate through the resin, each at a different rate. The plastic tubing on the left carries the emerging solution to test tubes mounted on an automatic sample changer.

A Spin-off in Space

The success of radiochemical detectors for weapons diagnostics led directly to the development of a device performing a similar function in quite a different environment—space. Radiation exposure of spacecraft and their crews was of course a concern of the nation's manned space-flight program, but measuring the kind and amount proved an awkward task. The instruments that do the job well in the laboratory require power, room, and attention that are at a premium in a spacecraft. Through previous collaborations on NASA space experiments, a member of the nuclear and radiochemistry group became aware of the problem and organized the designing of a radiochemical dosimeter that offered a partial but economical solution. Designed to measure neutron spectra and weighing approximately one pound, the dosimeter contained target specimens of uranium-238, yttrium-89, scandium-45, and titanium-46, -47, -48, -49, and -50. After a nine-day journey in space aboard the Apollo spacecraft that rendezvoused with the Soviet spacecraft Soyuz in July 1975, the dosimeter was returned to Los Alamos for radiochemical analyses within twenty-four hours after splashdown. The analysis of the radioactivities induced in the target isotopes, in conjunction with the body of neutron cross-section data derived from the group's experience with radiochemical detectors for weapons diagnostics, gave information on total neutron fluxes and on fluxes within various energy bins up to about 40 MeV. The data, apart from their intrinsic interest, were of value in interpreting other experiments carried out during the space mission. One noteworthy result was that the overall neutron exposures in the spacecraft had been lower by a factor of about 2.5 than was expected on the basis of calculations from previous flights. ■

—— Beta Decay ——▶

^{257}U → ^{257}Np → ^{257}Pu → ^{257}Am → ^{257}Cm → ^{257}Bk → ^{257}Cf → ^{257}Es → ^{257}Fm

↑   ↑

^{256}U → ^{256}Np → ^{256}Pu → ^{256}Am → ^{256}Cm → ^{256}Bk → ^{256}Cf

↑   ↑

^{255}U → ^{255}Np → ^{255}Pu → ^{255}Am → ^{255}Cm → ^{255}Bk → ^{255}Cf → ^{255}Es → $\boxed{^{255}\text{Fm}}$

↑   ↑

^{254}U → ^{254}Np → ^{254}Pu → ^{254}Am → ^{254}Cm → ^{254}Bk → ^{254}Cf

↑   ↑

^{253}U → ^{253}Np → ^{253}Pu → ^{253}Am → ^{253}Cm → ^{253}Bk → ^{253}Cf → $\boxed{^{253}\text{Es}}$

↑   ↑

^{252}U → ^{252}Np → ^{252}Pu → ^{252}Am → ^{252}Cm → ^{252}Bk → ^{252}Cf

↑   ↑

^{251}U → ^{251}Np → ^{251}Pu → ^{251}Am → ^{251}Cm → ^{251}Bk → ^{251}Cf

↑   ↑

^{250}U → ^{250}Np → ^{250}Pu → ^{250}Am → ^{250}Cm ^{250}Bk → ^{250}Cf

↑   ↑

^{249}U → ^{249}Np → ^{249}Pu → ^{249}Am → ^{249}Cm → ^{249}Bk → ^{249}Cf

↑   ↑

^{248}U → ^{248}Np → ^{248}Pu → ^{248}Am → ^{248}Cm

↑   ↑

^{247}U → ^{247}Np → ^{247}Pu → ^{247}Am → ^{247}Cm

↑   ↑

^{246}U → ^{246}Np → $\boxed{^{246}\text{Pu}}$ → $\boxed{^{246}\text{Am}}$ → ^{246}Cm

↑   ↑

^{245}U → ^{245}Np → ^{245}Pu → ^{245}Am → ^{245}Cm

↑   ↑

^{244}U → ^{244}Np → $\boxed{^{244}\text{Pu}}$

↑   ↑

^{243}U → ^{243}Np → ^{243}Pu → ^{243}Am

↑   ↑

^{242}U → ^{242}Np → ^{242}Pu

↑   ↑

^{241}U → ^{241}Np → ^{241}Pu → ^{241}Am

↑   ↑

^{240}U → ^{240}Np → ^{240}Pu

↑   ↑

^{239}U → ^{239}Np → ^{239}Pu

↑   ↑

^{238}U ^{238}Np → ^{238}Pu

◀—— Neutron Capture ——

Postulated mechanism for production of heavy isotopes observed in the debris of the first large-scale thermonuclear explosion, the Mike test of 1952. (Known isotopes are in white type; those identified for the first time in the Mike test are also outlined in white. Isotopes as yet unidentified are in black type.) In the high-neutron-flux environment of that explosion, uranium-238 nuclei climb the mass ladder rapidly (in less than a microsecond) by successive neutron-capture reactions, with many reaching the lower rungs and few reaching the higher rungs. Then, on a slower time scale the neutron-rich uranium isotopes begin a migration toward stability by successive beta decays. In a beta-decay chain (represented here by a left-to-right row of constant-mass isotopes) the half-lives of the members tend to increase—from fractions of a second to months or years—with progress toward "beta stability." Also contributing by a similar mechanism to production of the observed heavy isotopes are neptunium isotopes formed during the explosion by reactions such as $^{238}U(^{2}H,n)^{239}Np$. It is believed that the neptunium isotopes are the predecessors of most of the higher mass products.

out to be a proverbial gold mine for basic nuclear science as well as for the science and technology of thermonuclear explosions. We return to this exciting subject in a later section.

The remaining years of the '50s up to the moratorium on testing of November 1, 1958 were a period of hectic activity and vigorous scientific growth for the group. During this period there were four weapons test series at Nevada and three in the Pacific, for all of which the group provided essential diagnostic information. In addition, research on radiochemical procedures and on nuclear properties proceeded along several lines, some of them prompted by the needs of weapons diagnostics, others by ideas, special sources, and facilities deriving from the test activities. An especially important factor in this work was the availability of state-of-the-art radiation measuring equipment, including 4π beta counters, magnetic discrimination instruments for positron counting, multichannel pulse-height analyzers for alpha and gamma scintillation spectrum measurements, and a magnetic beta-ray spectrometer.

Diagnostics for Nuclear Rocketry

The middle '50s saw another significant change for the nuclear and radiochemistry group: R. W. Spence, who had created the group in late 1946 and led it through nine years of expansion, was appointed second in command of a new division set up to study nuclear rocket propulsion, and his place as group leader was taken by G. A. Cowan. An early member of the Manhattan Project at Princeton University and the University of Chicago and with Los Alamos for Operation Crossroads, Cowan had been with Spence's group since 1949 until he was brought to the J Division office in early 1955. Shortly after assuming the group leadership in November 1955 (the group was then known as J-11), he initiated what was to become a second major program for the group over the next fifteen years: the diagnosis of nuclear rocket per-

formance by radiochemical techniques.

The program to investigate the use of nuclear reactors for rocket propulsion, the Rover program, represented a major new initiative for the Laboratory. The basic concept was to heat a low-molecular-weight gas—hydrogen was the obvious choice—by passing it through a compact reactor and to direct the now high-temperature gas stream through a nozzle as a high-specific-impulse propellant. Practical exploration and development of this concept involved diagnostic needs for which radiochemical techniques were well suited: measurement and calibration of fission-energy release, analysis of fission distribution in the reactor fuel elements, assays of the amount of fuel and fission products escaping from the reactor into the propellant, and a variety of problems associated with creation and migration of radioactivity under high-temperature conditions.

For a reactor with uranium-impregnated graphite fuel, the design type provisionally selected for Rover, there were three concerns of special importance to high-temperature operation: the loss of uranium fuel by diffusion out of the fuel elements, the loss of delayed-neutron-emitting fission products that provide the margin of reactor control, and the escape of fission products in general to the propellant stream. With these concerns foremost in mind, the group began immediately an experimental study, in the temperature range expected for the reactor, of the rates of diffusion and volatilization into a gas stream of a large number of elements from fission of uranium in a graphite matrix. Although some data were available from previous studies conducted elsewhere, it was necessary to have a wider and self-consistent data base suited to the Rover components. From the large amount of these data three important results emerged: none of the uranium diffused out; although substantial amounts of the delayed-neutron emitters bromine and iodine were lost at temperatures of 2000 degrees Celsius

and above, more than half of the delayed-neutron species were retained; and some of the fission-product elements, such as silver and cadmium, were found to be very mobile. In one experiment the sharp variation in mobility was exploited to measure the half-life of palladium-115, a newly discovered fission-product isotope. Uranium-loaded graphite slugs were irradiated with neutrons, allowed to decay at room temperature for varying lengths of time, and then heated to expel the more volatile products, among them being the cadmium-115 granddaughter of palladium-115. (This procedure is somewhat analogous to a chemical "milking" experiment.) Analyses for the cadmium-115 showed the half-life of palladium-115 to be 47 ± 3 seconds.

The studies of the variations in diffusion rates with element and temperature led also to the development of a means for indirect determination of temperature distribution in the reactor. Small cartridges of graphite loaded with radioactive tracers selected to cover a range of diffusion rates were emplaced at various locations in the reactor during assembly. These "radiochemical thermometers" were recovered after a reactor test run and analyzed for tracer content. Comparison of the tracer losses with losses from identical units put through calibration runs in ovens back at the laboratory gave the approximate temperatures at the reactor locations. Our records show that in a typical full-scale reactor test we employed sixty such thermometers, each one loaded with the tracers barium-133, americium-241, and plutonium-239, listed in order of decreasing diffusivity.

By far the largest component of the nuclear and radiochemistry group's participation in the Rover program was the detailed post-mortem of the fuel component of the various reactors after full-scale tests. The reactors were disassembled at the Nevada Rocket Development Site, and the fuel elements were returned to Los Alamos for determination of fission distribution, total
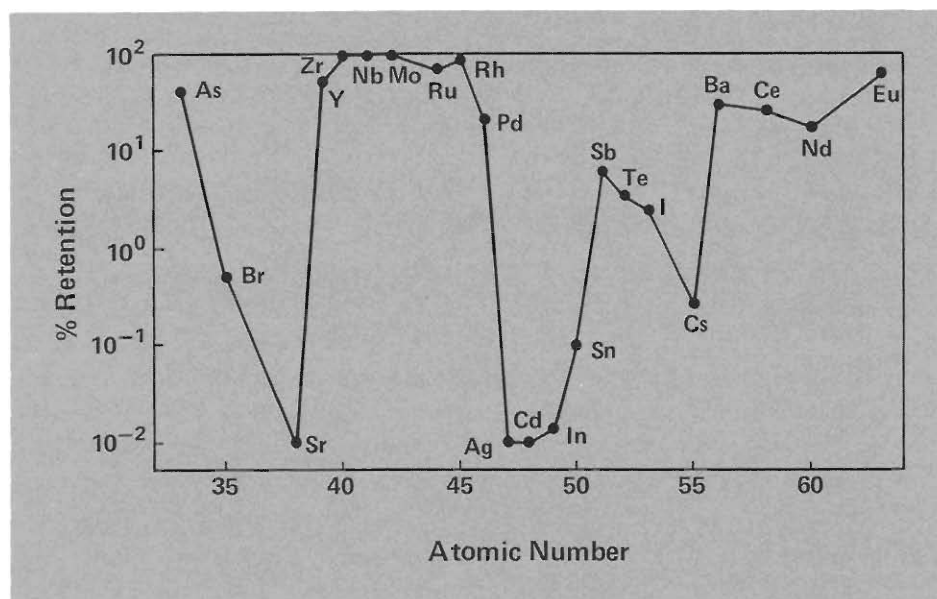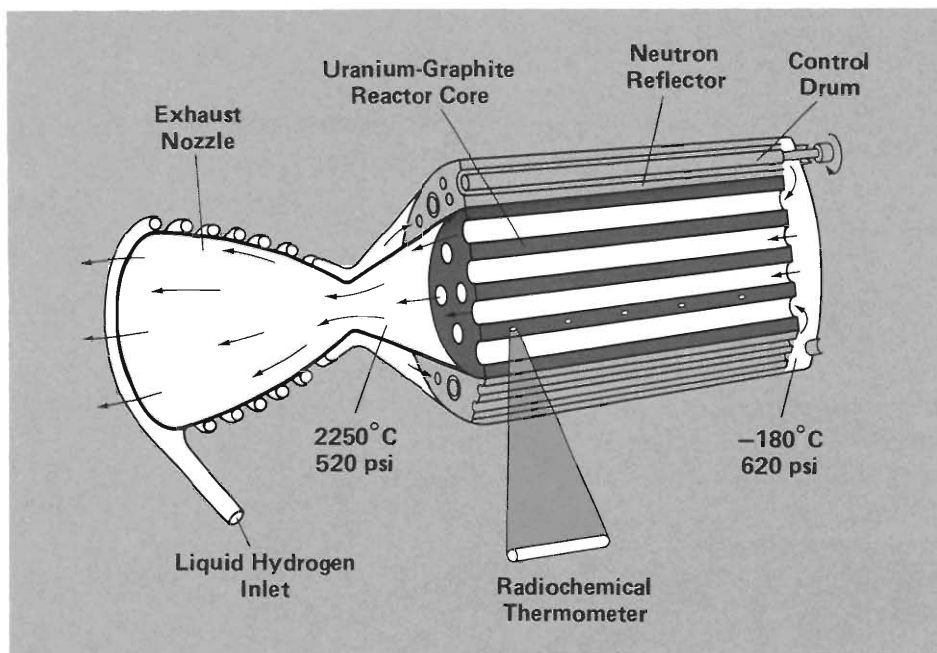
From 1955 to 1970 Los Alamos was investigating the principles of nuclear rockets, rockets propelled by a low-molecular-weight gas heated to high temperature by passage through a nuclear reactor. The upper figure shows schematically the design of one of the rocket engines built and tested by the Laboratory. [From R. W. Spence, International Science and Technology, 58-65 (July 1965).] The hydrogen not only provided the rocket's thrust as it exited through the exhaust nozzle but also cooled the nozzle as it entered the system in liquid form through the coiled pipe. The Laboratory's nuclear and radiochemistry group was called upon to study various aspects of rocket reactor behavior, including the diffusion of fission products from the graphite fuel matrix at the expected operating temperatures. The lower figure shows data from one such study in which members of the group measured the amounts of various fission-product elements remaining in a graphite sample after it was held at 2400 degrees Celsius for 4 minutes. The variation of diffusivity with element and with temperature led to the development of "radiochemical thermometers" for determining temperatures within the reactor core during a test run. The thermometers contained radioactive isotopes of elements covering a range of diffusivities. The losses of the isotopes measured after the test run gave the temperatures at various reactor locations.

fission, and fission-product loss to the graphite moderator and to the atmosphere. This large-scale effort, extending from 1958 to 1967, had important consequences for future undertakings of the group. It brought to the radiochemistry site a versatile chemical hot-cell facility and a complement of up-to-date radiation-measuring apparatus with computerized data-processing equipment (the latter included a PDP-8 computer acquired in 1966). Even more important than the facilities, it brought the group valuable experience and a broadened outlook in state-of-the-art radiochemical operations on the 100-curie scale.

Scientific Applications of Nuclear Explosions

In the middle and late 1950s the nuclear chemistry group undertook another activity that was to make a significant contribution to basic nuclear science: the specific application of nuclear explosions to the measurement of quantities and the production of materials that lay outside the reach of conventional laboratory facilities. This research direction, initiated and actively promoted by Cowan, had its first noteworthy success with the "wheel experiment" carried out during a nuclear weapon test in 1961. This experiment was aimed at measuring the variation in mass symmetry of uranium-235's fission at its neutron resonances. [A little explanation may be in order here: for the neutron-induced fission of, say, uranium-235, the term "mass symmetry" describes the extent to which the compound nucleus uranium-236 (the intermediate product formed by absorption of a neutron) splits nearly in half, and the term "neutron resonance" describes the enhanced absorption of neutrons with energies corresponding to the differences between that of uranium-235 and those of semistable states of the compound nucleus.] A disk faced with uranium-235 was set spinning in front of a slit at the upper end of a vacuum pipe leading to an underground
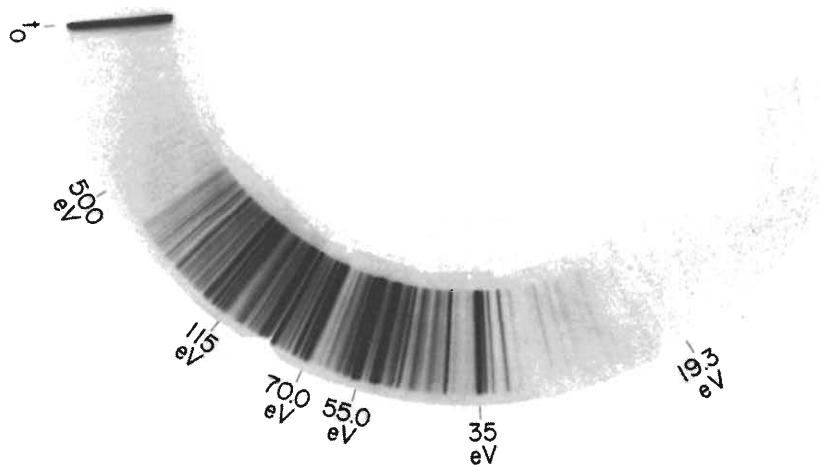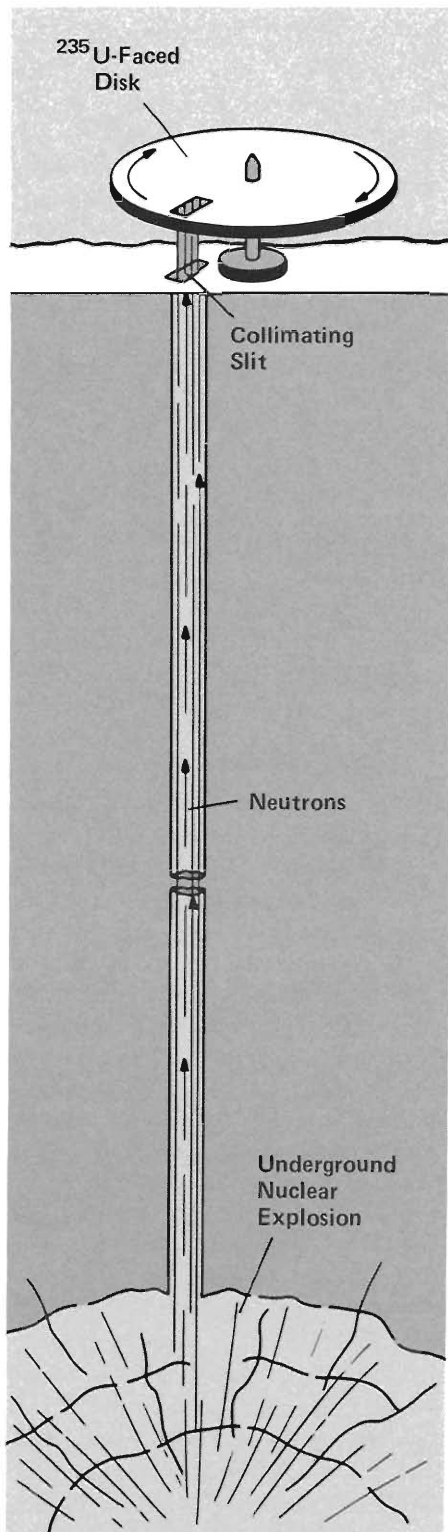
Shown schematically at left is the setup for the "wheel experiment" to determine the variation in mass symmetry of the fission induced in uranium-235 by neutrons with energies corresponding to its neutron resonances. The neutron source was an underground nuclear explosion, which produces, essentially in a single burst, neutrons with speeds ranging from one-fifth to ten millionths the speed of light. By the time the neutrons reach the target disk, they have been separated in energy by time of flight; that is, neutrons with different energies reach the target at different times—the fast first, the laggards later. Because the disk is rotating, the neutrons arriving at different times strike the target at different locations. Those neutrons with resonant energies induce many fissions and create bands of fission products in the uranium-235. Shown above is an autoradiograph of the fission products on the disk's surface. The energy scale was derived by identifying the time of the explosion t_o (revealed on the disk by a large number of fast-neutron-induced fissions) and a few prominent resonances.

nuclear explosion. The pulse of neutrons from the explosion, separated in energy by time of flight, created in the uranium-235 a series of fission-product bands corresponding to the resonances. Radiochemical analysis of the individual bands for total fissions and for silver-111 and cadmium-115, products of nearly symmetric fission, gave the symmetric-fission yields at the resonances. This and subsequent experiments with improved techniques showed that the symmetric-fission yields fall into two groups averaging 0.59 and 1.11 times the symmetric-fission yield for fission induced by thermal neutrons. The resonances with the lower and the higher symmetric-fission yields, respectively, could be identified with states in uranium-236 with total angular momentum J of 4 and 3. Similar results were obtained for the resonances of plutonium-239, but in this case the higher and lower symmetries are associated with the $J = 1$ and the $J = 0$ states of plutonium-240.

Another scientific application of nuclear explosions was one that had been brought to light in startling fashion with the Mike test: the production of very heavy nuclides. The results of that test had given rise to a wave of hope amongst some of the heavy-element research fraternity. They reasoned that since
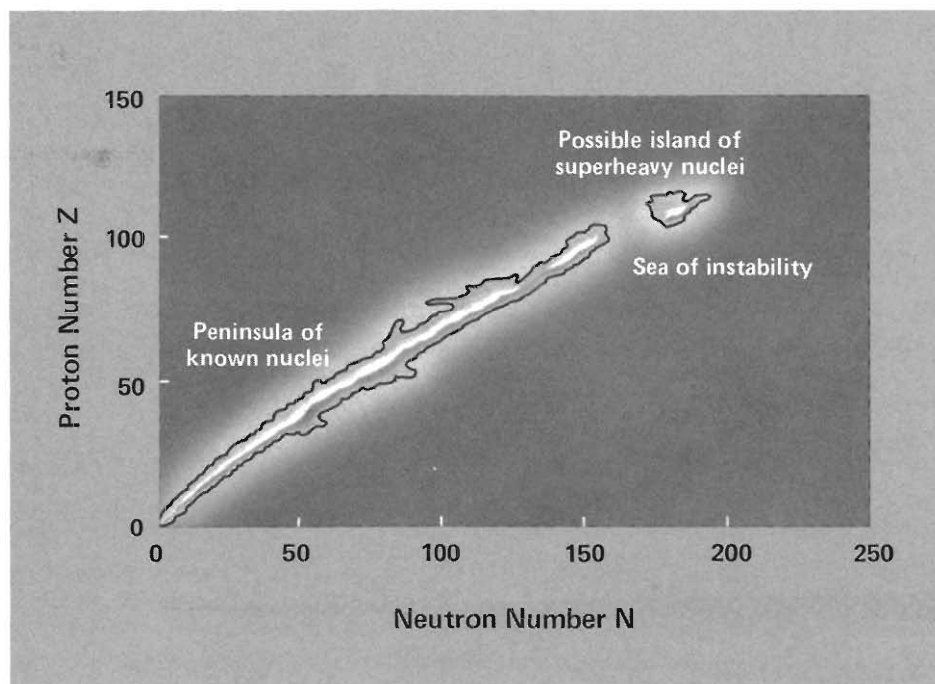
Attempts to reach a region of predicted semistable nuclides in the vicinity of Z = 114 and N = 184, the "island of stability," have occupied nuclear scientists since the '50s. According to current thinking, the best prospect of reaching it lies in single-step collisions, with minimum excitation, between neutron-rich projectiles and neutron-rich targets—in effect a quick, shallow underwater trip to the island. (Adapted from computergraphics by Ray Nix.)

the Mike device was designed to achieve other technical goals—the production of heavy elements was, after all, only incidental—it did not seem unreasonable that other devices of this kind could be modified to achieve even higher instantaneous neutron fluxes and thus provide a path to synthesis of still heavier elements and possibly even the superheavies, the predicted semistable nuclides in the vicinity of the doubly closed-shell nuclide $^{298}114$. (The stability of nuclides with approximately 114 protons and 184 neutrons is predicted by the shell model of the nucleus. This model explains the observed stability of nuclides with "closed" shells containing certain "magic" numbers of protons or neutrons. Other examples of

doubly closed-shell nuclides are calcium-40 and lead-208.) Moreover, by the '60s there was reason to believe that the high neutron fluxes could be achieved in explosions producing much less radioactivity and releasing much less energy, say of the order of a few kilotons. Between 1962 and 1969 several sophisticated devices modified for production of heavy elements were actually fired in underground tests in Nevada, some by Los Alamos and some by Livermore. Although radiochemical examination of the products of these tests turned up valuable nuclear information, it also clearly brought to light a fundamental limitation on heavy-element synthesis—spontaneous fission. The fluxes achieved in these later tests were high, but

the rapidly increasing probability of spontaneous fission with increasing isotopic mass set an effective limit on the masses of the products attainable. The heaviest product recovered was fermium-257, with a half-life of 100 days. [The heavier isotope fermium-259, discovered in 1976 by a Los Alamos/Livermore team employing the $(^3H,p)$ reaction on a fermium-257 target, decays by spontaneous fission with a half-life of only 1.5 seconds.] There has been some speculation on the possibility of reaching the predicted "island of stability" near $^{298}114$ by irradiating suitable heavy-element targets in a very high-flux nuclear explosion (or perhaps one bootstrapped on another) to make an "end run" around the zone of nuclides prone to spontaneous fission, but the prospects do not at present appear sufficiently promising to warrant a test. Meanwhile, this heavy-element work has indicated that a large area on the neutron-rich side of the isotope chart represents nuclei known to have been produced (or to be producible) in nuclear explosions but remaining undescribed and currently unavailable by any other known means. Here the limitation lies not in the technology of thermonuclear explosives but in the technology of sample recovery. Development of the means to retrieve and partially process samples from an underground explosion within five or ten minutes of the event could open up a large new field of nuclear studies.

Tools Old and New

In science it is axiomatic that new advances follow the introduction of new tools and new techniques. Pressed by insistent demands for accurate and timely diagnostic data, the group and its instrumentation czar J. P. Balagna had early on recognized the necessity for first-rate equipment. New tools of the trade were tested and incorporated into the group's instrumentation complex as soon as they appeared on the scene. A particularly useful addition was an isotope

separator purchased in 1964. With it we could separate interfering isotopes of various elements and thus perform for the first time accurate analyses for second- and third-order (and in some cases even higher order) products of neutron reactions. This capability significantly broadened the range and utility of the radiochemical detector technique for weapons diagnostics. The isotope separator has been used as well for experiments in basic nuclear science, including a tour de force involving tightly scheduled operations at Oak Ridge, Los Alamos, and the Nevada Test Site in which the fission cross section of 6.75-day uranium-237 was measured at neutron energies from 43 eV to 1.83 MeV.

Semiconductor radiation detectors, silicon units for charged particles and soft x rays and lithium-drifted germanium units for gamma rays, also became available to the group in the '60s. With their fifty-fold increase in energy resolution over the then standard sodium iodide scintillation detectors, semiconductor detectors made it possible to identify and precisely assay individual radionuclides in mixtures and thereby permitted us to redirect a great deal of radiochemical processing and counting time to new measurements. At Los Alamos as elsewhere, the new detectors also brought about a jump in the detail and range of information to be obtained from basic nuclear research.

There has been another component of the group's facilities and capabilities that has expanded its interests to areas not commonly associated with nuclear and radiochemistry. Ever since the first atmospheric tests in Nevada in 1951, explosion-cloud sampling had been performed by the U.S. Air Force with manned aircraft; the actual cloud sampling was directed at close range by a scientist from the group serving as a crew member in the sampling squadron. With the cessation of atmospheric testing in 1963, the sampling director, some of the aircraft, and, most important, the accumulated experience

became available for other studies. One of the first studies involved flights through the discharge plumes of the Rover reactor tests to provide information on the radioactivity carried out by the propellant gas. Then, in the middle to the late '60s, the rapidly increasing national concern over environmental pollution steered the sampling efforts toward investigation of airborne pollutants. The aircraft sampling facilities were used to survey and measure forest-fire clouds, industrial smokestack discharges, and regional air quality. By microscopic examination, neutron-activation analysis, and other specialized techniques group members were able in many cases to establish telltale "fingerprints" of individual sources by which their contributions to atmospheric pollution could be tracked far from the point of origin.

The mid '70s brought to the nuclear and radiochemistry group a state-of-the-art mass spectrometry facility that made possible quantitative measurement of isotopes by direct counting of atoms. Its development represented the convergence of two trends, one in supply and one in demand. The demand lay in the pressure to extract more detailed and sophisticated diagnostic information from the debris of nuclear weapons tests. On the supply side, steady improvement in the apparatus and techniques available to mass spectrometry was bringing it within reach as a cost-effective adjunct to the diagnostics program. The most common diagnostic task requiring mass spectrometry is determining the isotopic compositions of the uranium and plutonium fuels remaining in the debris. From such data is obtained the fraction of the fissile fuel consumed by nonfission reactions. But, as in other pursuits, with every boon comes a burden. Mass spectrometers are notoriously finicky, and every sample must be meticulously prepared to free it of all other elements, real or suspected, that come with it in the raw debris. Still, the effort spent in setting up the facility and getting it to work as it should has begun to pay off. The mass spectrometrists

now not only supply essential isotope data on fissile elements and on radiochemical detector elements but also have made possible some new initiatives in basic research.

One quite recent initiative that demands the capabilities of the mass spectrometry facility is the use of "heavy" methane molecules as tracers to study atmospheric circulation and mixing patterns. ("Normal" methane contains the common isotopes carbon-12 and hydrogen-1; in the heavy methanes these isotopes are replaced by the rare but stable isotopes carbon-13 and/or hydrogen-2.) In a 1980 trial experiment carried out in collaboration with the National Oceanic and Atmospheric Administration, heavy methanes were released at Norman, Oklahoma, and their concentrations were measured in samples collected at a network of stations and by the U.S. Air Force. Evidence of the heavy methanes was found even at East Coast locations. Since the samples contained much larger concentrations of normal methane, state-of-the-art mass spectrometry was essential to the experiment. The mass spectrometers currently in use at Los Alamos can measure about one part of methane-20 in a billion parts of normal methane and one part of methane-21 in 200 billion parts of normal methane. (These discrimination capabilities may be increased by a factor of a thousand with new techniques involving sophisticated particle-identification apparatus at the Laboratory's vertical electrostatic accelerator.) The success of the trial experiments has led to preparations for further experiments on a continental scale.

Members of the group also, of course, have access to other Laboratory facilities of great value for their programmatic and research interests. Among these facilities, until it was shut down in 1974, was the old cyclotron brought here during World War II, which provided beams of low-energy charged particles. This cyclotron was the world's first practical source of "lightweight

alpha particles," or helium-3 ions, and nuclear chemists had measured excitation functions (cross section dependence on energy) of the reactions between helium-3 and target nuclei ranging from carbon to uranium.

Another valuable facility, the vertical electrostatic accelerator/FN Tandem Van de Graaff complex, came on line in 1964; it was the first to offer accelerated beams of tritons with energies up to more than 20 MeV. With these tritons nuclear researchers could study nuclei by reactions such as $(^3H,p)$, $(^3H,\alpha)$, and $(^3H,\alpha p)$. Also available were high-energy neutrons [from the $^2H(^3H,n)^4He$ reaction] for measuring excitation functions of (n,xn) reactions at neutron energies well above 14 MeV, a regime of considerable interest to the weapons program. This accelerator complex has served as the focus for a nuclear chemistry/nuclear physics (the boundary line is sometimes indistinct) collaboration on researches of frontline interest: the fission process in a wide range of nuclei and excitation, the properties of the heaviest nuclei, and the processes occurring when complex nuclei are bombarded with other complex nuclei.

In addition, the charged particles available from the Van de Graaff provide a way—often the only way—to study neutron reactions of isotopes important to radiochemical weapons diagnostics. As an example, consider the isotope yttrium-86, which is among the neutron-deficient yttrium isotopes formed by (n,xn) reactions when yttrium-89, the common stable isotope of yttrium, is employed as a radiochemical detector in a nuclear device. Analyses of debris samples for the various yttrium isotopes, together with the known (n,xn) cross sections, are expected to provide values for the flux of neutrons from the thermonuclear fusion of deuterium and tritium. But the flux determination is complicated by the fact that some of the yttrium-86 is consumed by further neutron reactions, such as (n,γ), (n,p), and (n,np). To correct for this complication, information about the cross sections of these parasitic reactions is needed but is difficult or

impossible to obtain directly. The cross section for, say, the (n,p) reaction can be determined indirectly, however, by creating excited states of yttrium-87 [the intermediate product of the (n,p) reaction] and measuring the fraction of these excited nuclei that decay by proton emission. The excited yttrium-87 nuclei are produced through the reaction $^{86}Sr(^3He,^2H)^{87}Y$ by employing helium-3 ions from the Van de Graaff and a target of strontium-86 (an uncommon but stable isotope available from Oak Ridge). This example illustrates that by using well-known nuclear physics strategies we can circumvent the difficulties sometimes arrayed against us when we try to study a nuclear reaction as it actually occurs.

The Laboratory's reactor complex, in particular the series of high-neutron-flux installations (of which the Omega West reactor and the Pajarito Site critical assemblies are the current representatives), has also been an essential and versatile resource for the nuclear and radiochemistry group. Nuclear explosive devices are essentially one-shot neutron reactors specially designed to achieve very rapid power buildup and energy release, and the great variety of radiochemical procedures needed to diagnose their performance has to be developed and calibrated with more conventional neutron sources.

Mentioned last only because of its more recent vintage, the Laboratory's meson physics facility (LAMPF) has considerably broadened the scope of the group's nuclear chemistry research. We elaborate on this topic in a later section.
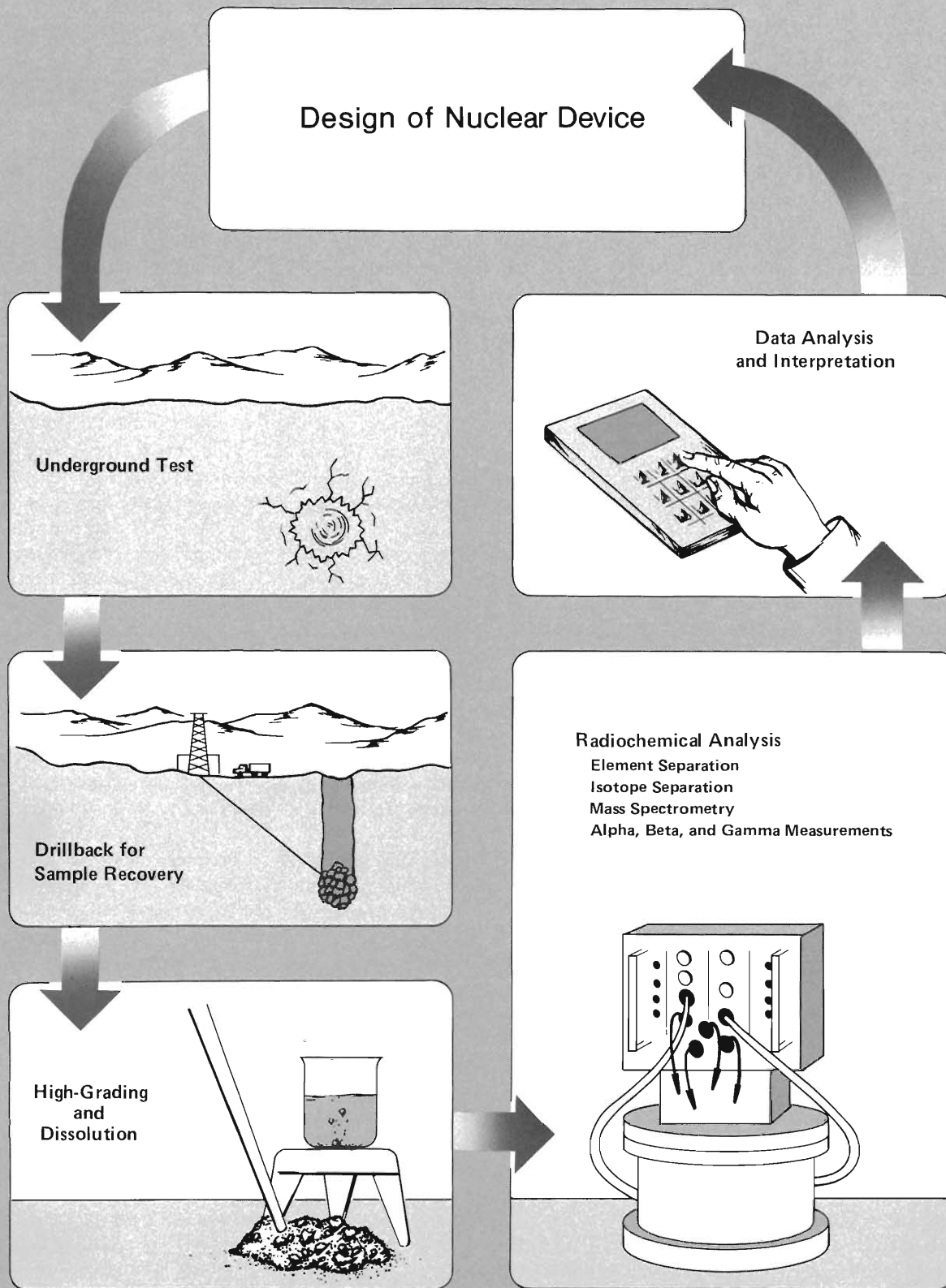
From Underground Testing to Isotope Geochemistry

The Limited Test Ban Treaty of 1963, which restricted all nuclear test explosions to underground, had a far-reaching effect on the course of nuclear and radiochemistry at Los Alamos. Compared with atmospheric testing, underground testing had both draw-

backs and advantages for radiochemical diagnostics. Access to the debris was slower—days compared to hours—and the material of interest in the debris had to be extracted from large amounts of fused earth, usually containing unwanted products from neutron activation of trace elements in the earth. But for most diagnostic purposes the benefits more than compensated for the drawbacks. The debris stayed where it was made, available for recovery in large amounts at any time and with simpler control of personnel radiation exposure.

The experience and expertise gained in dealing with the products associated with underground nuclear explosions contributed to some noteworthy accomplishments and new initiatives. For example, starting with 85 kilograms of bastnäsite ore from a rare-earth mine near Mountain Pass, California, D. C. Hoffman and her colleagues succeeded in isolating a plutonium fraction that, when subjected to mass-spectrometric analysis at the General Electric Research Laboratory, showed the presence of plutonium-244. (Bastnäsite is a rare-earth mineral whose formation should have concentrated also any primordial plutonium.) Reported in 1971, this discovery of plutonium-244 in nature had significant consequences for our ideas about nucleosynthesis and the distribution of elements in the solar system. In other high-sensitivity measurements another staff member and his postdoctoral associate, in collaboration with associates at Livermore and Hanford, obtained evidence for niobium-92 and -94 in natural niobium. These investigations were the precursors of what is today a large and far-ranging research effort in isotope geochemistry.

The group's capabilities in applying radiochemical techniques to measuring very minute quantities of elements in earth samples, coupled with the interests of group member C. J. Orth, led recently to the discovery of evidence in support of a controversial explanation for the extinction of many plant and animal species, including the dinosaurs, that

Design of Nuclear Device

Underground Test

Drillback for Sample Recovery

High-Grading and Dissolution

Data Analysis and Interpretation

Radiochemical Analysis
- Element Separation
- Isotope Separation
- Mass Spectrometry
- Alpha, Beta, and Gamma Measurements

Radiochemical diagnostics provide essential feedback to nuclear weapon design. A variety of analytical techniques are called upon to produce the necessary range of information about a weapon's performance during a test explosion.

marks the boundary between the Cretaceous and Tertiary geologic periods. A Berkeley team headed by Luis and Walter Alvarez proposed that an interruption of the food chain was responsible, an interruption caused by a blotting out of the sun by debris from the impact of an extraterrestrial object. They based this hypothesis on unusually high abundances of iridium found in marine sediments from Italy at a stratum coincident with the Cretaceous-Tertiary boundary. (Iridium is a platinum-family element exceedingly scarce in the earth's crust and somewhat less scarce in extraterrestrial matter.) Similar iridium "anomalies" were subsequently discovered at other locations, but all were found in marine sediments and could possibly be attributed to concentration by ocean processes. Orth and his radiochemistry team members, in collaboration with experts in Cretaceous stratigraphy and plant life from the U.S. Geological Survey, selected a likely drill site in northern New Mexico and succeeded in discovering, in *freshwater* sediment, a strong iridium concentration spike that correlated exactly with the Cretaceous-Tertiary boundary as defined by changes in abundances of pollen species. Since the peak concentration of iridium in normal earth samples is only parts per trillion, very sensitive analytical techniques involving neutron activation and radiochemical separation were required to detect the anomaly. This research area is fast moving, and we expect future developments to broaden the scope of our contributions still further.

The Winds of Change

The period from the mid '60s to the early '70s was a time of transition for the Laboratory, a transition from concentration of effort on nuclear weapons to participation also in a broad spectrum of other high-technology topics of national concern. Nuclear and radiochemistry entered a corresponding transition. Significant new re-

search was initiated on nuclear structure and reactions (at the electrostatic accelerator complex and at LAMPF), on the application of new radioisotopes to medicine and of stable isotopes to atmospheric studies, in isotope geochemistry and cosmochemistry, and on the migration and containment of nuclear wastes. Discussion of research in the latter area has been reserved for another article in this issue of *Los Alamos Science.*
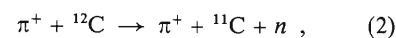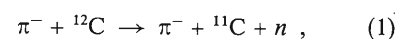
Linked with the broadening range of research was a sequence of leadership changes following the appointment in 1970 of Harold M. Agnew as the third Director of the Laboratory. Group Leader Cowan was made Leader of the new Chemistry-Nuclear Chemistry Division, which was created to incorporate research in inorganic, physical, and nuclear chemistry. The nuclear and radiochemists under newly appointed Group Leader James E. Sattizahn became CNC-11.
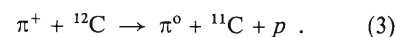
Nuclear Chemistry at LAMPF

With the start-up of the Clinton P. Anderson Meson Physics Facility in the early 1970s, the chemists found new horizons opened to them in both applied radiochemistry and basic nuclear science. This 800-MeV linear proton accelerator and its array of auxiliary equipment, built to produce beams of protons and pi mesons (pions) at intensities several hundredfold greater than had been available anywhere else before, brought within reach a wide range of phenomena previously almost inaccessible to practical measurement. One of these phenomena is the interaction of pions with nuclei, and small teams of nuclear chemists began to study pion-induced nuclear reactions on targets ranging from carbon to uranium by measuring the radioactive products. (We note here a commonly used distinction made a little less inaccurate by use of the qualifier "many": many nuclear *chemists* study nuclear reactions by measuring radioactive products remaining in a target *after* bombardment; many nuclear

physicists study nuclear reactions by measuring particles emitted from a target *during* bombardment.)

An interesting example is the triplet of pion-induced reactions that produce carbon-11 in a natural carbon (predominantly carbon-12) target:

$$\pi^- + {}^{12}C \rightarrow \pi^- + {}^{11}C + n , \qquad (1)$$

$$\pi^+ + {}^{12}C \rightarrow \pi^+ + {}^{11}C + n , \qquad (2)$$

and

$$\pi^+ + {}^{12}C \rightarrow \pi^0 + {}^{11}C + p . \qquad (3)$$

Once the cross sections for these reactions are known at various incident pion energies, they become good intensity monitors for negative and positive pion beams. One places a carbon-containing target in the pion beam for a few minutes, withdraws it, and measures the carbon-11 produced; a simple calculation gives the number of pions that struck the target. These reactions contain a basic physics interest as well. Examination of reactions 1 and 2 suggests that the incoming pion, whether negative or positive, simply hits a single neutron in the carbon-12 nucleus and knocks it out; reaction 3 is the same except that the positive pion passes its positive charge to a neutron during the collision and a proton emerges. Now it is considered as firmly established in nuclear physics that, at pion kinetic energies around 180 MeV (the so-called 3/2,3/2 resonance), the probabilities for the elementary reactions $\pi^- + n \rightarrow \pi^- + n$, $\pi^+ + n \rightarrow \pi^+ + n$, and $\pi^+ + n \rightarrow \pi^0 + p$ should be as 9 to 1 to 2. Thus, if a carbon-12 nucleus is merely a collection of neutrons and protons, the ratio of the cross section for reaction 1 to that for reactions 2 and 3 together should be 3. (When carbon-11 is measured, reactions 2 and 3 are indistinguishable.) When carefully measured by the nuclear chemists, however, the ratio came out to be not 3, but 1.6. This seemingly simple discrepancy presented the theoreticians with an interesting problem, and they

have devised a model that solves it. The key to their solution lies in taking into account that, although reactions 1, 2, and 3 as we have written them may represent what happens *at the site of a collision inside a nucleus,* the emerging neutron (proton) is likely to hit and trade places with a proton (neutron) while on its way out of the nucleus and produce another mass-11 nuclide, boron-11 (nitrogen-11). Thus, the carbon-11 nuclei that the chemist sees may be fewer than the total that were initially produced in the collision. They represent the cases where "nothing went wrong."

Another research interest of the nuclear chemists at LAMPF concerns the "exotic" nuclei produced when 800-MeV protons strike nuclei, especially heavy ones. This interaction yields a wide assortment of particles and nuclear fragments ranging from pions and neutrons to nuclei only a little lighter than the target. Among these fragments are nuclei, many of them still unidentified, that represent the extremes of what can exist at all. Some of them have a great excess of neutrons, so great that their last few neutrons are barely held; these are described as almost neutron-unbound or near the "neutron drip line." There exist other nuclei at the opposite extreme: severely deficient in neutrons and rich in protons, that is, almost proton-unbound, or near the "proton drip line." These exotic nuclei are hard to find. Not only are they produced very rarely in nuclear reactions, but they have half-lives so short (seconds or less) that they cannot be isolated and identified by conventional radiochemical techniques. A team of nuclear chemists devised a method to identify some of these exotic products in flight, that is, as they emerge from the proton/heavy nucleus reaction. Measuring a product's time of flight from the production site to a detector gives its velocity; measuring its kinetic energy as it is stopped in the detector then gives its mass from the expression $E = \frac{1}{2}mv^2$ (the correction for rel-

The Discovery of Thorium-236

A bout a year before LAMPF's experimental program was officially launched with the production of the first mesons in 1973, the nuclear chemists conducted an experiment, Experiment Zero, that led to the first scientific discovery and publication from this new installation. By bombarding a target of uranium-238 with the high-intensity 100-MeV proton test beam that had just become available, they produced a new heavy isotope of thorium, thorium-236. With short bombardments followed by fast radiochemical isolation and electromagnetic isotope separations, they were able to identify and characterize this previously unknown isotope. It decays by beta emission with a half-life of 37.5 minutes. ∎

ativistic effects is small); and measuring its rate of energy loss in the detector assembly gives its nuclear charge. Thus, by electronic means one establishes the identity of the isotope without chemically separating it or measuring its decay. In a series of experiments using thin targets of uranium and nickel, the nuclear chemists have identified five new examples of neutron-rich exotic isotopes: neon-27, magnesium-31, magnesium-32, aluminum-34, and phosphorus-39. (The heaviest stable isotopes of these elements are neon-22, magnesium-26, aluminum-27, and phosphorus-31.) Others are currently under study.

Present nuclear theory cannot predict accurately the boundaries of nuclear existence, and there is now evidence that even the exotic nuclei named above may be short of the neutron drip line. To illustrate this point, we pick an example from low-atomic-number nuclei measured elsewhere. The element beryllium, with an atomic number of 4, has a single stable isotope, beryllium-9. The question has been asked, "What is the heaviest possible isotope of beryllium?" A team at the University of California Bevatron made a search, employing an experiment similar to

the one described above. At the upper end of the mass scale they found evidence for beryllium-11, -12 (both already known), and -14, but not for beryllium-13, -15, or -16. Thus, it appears that the neutron drip line occurs beyond beryllium-14 and that beryllium-16 is probably neutron-unbound. On the other hand, beryllium-11 is probably the heaviest odd-mass beryllium isotope that is neutron-bound. The difference between the odd- and even-mass isotopes is not surprising to nuclear scientists; it reflects the well-known preference of nuclear particles to occur in pairs.

So far we have talked in terms of merely establishing the existence of exotic nuclei. The data would be much more valuable if they told in addition *how stable* such nuclei are. The Los Alamos team is now at work on a new apparatus that should provide this kind of information. It will still measure time of flight, but with sufficient precision to give a spectroscopically useful isotope mass, which is a direct measure of nuclear stability. When operational, the instrument should yield masses on at least thirty exotic nuclei with accuracies of 200 keV or better, a figure that should lead to a new kind of thinking

about these nuclei.

After the proton beam at LAMPF has passed through the last of the particle-generating targets, it is considered to have done its work. Attenuated to between 50 and 75 percent of its starting intensity and too diffuse for normal use, it is refocused slightly and sent to the "beam dump." But even in its death throes it can produce large amounts of radioactivity in the materials in its path. Inserting targets of their own into the beam-dump zone by means of a remotely controlled facility specially engineered for the purpose, radiochemists obtain a variety of radioisotopes that they isolate and prepare for experiments in nuclear medicine. A separate radiochemistry group has been built up to explore and develop this family of applications.

Another offering on the LAMPF menu, negative muons, has provided the chemists with a new probe of matter whose utility and applications they are still exploring. (A negative muon, loosely speaking, is a semi-stable, heavy version of an electron. These particles are available at LAMPF from the decay of negative pions.) Experimentally, one places a specimen of interest in a beam of negative muons and measures the intensities of the muonic x rays emitted as the muons captured by individual atoms de-excite through their levels in the atoms. Each element is identifiable by its set of muonic x rays, just as it is by its set of ordinary electronic x rays. The aspect of this phenomenology that attracts the attention of the chemists is one first observed over two decades ago: the muonic x-ray intensity patterns of the elements in a compound and the relative number of muons captured by the individual elements in a compound *vary with the compound's structure and state.* Thus, if one could learn something about the nature of this variation, one might use it as a tool to investigate chemical structure.

Beginning with an experimental program

that got under way in the mid '70s, nuclear chemists have studied the intensity effects on a variety of target materials chosen to elicit chemical structure factors. Along the way they have turned up a number of curious results, some of which are beginning to make sense in terms of the chemical bond. For example, the relative probability of capture of a negative muon by the two elements in a binary compound had been expected to vary approximately as the atomic numbers of the two elements weighted by their stoichiometric abundances; thus, in boron nitride (BN) the ratio of the capture probabilities $c(B)$ and $c(N)$ would be 5/7, or 0.714. Actual measurement, however, gave a ratio of 0.258 ± 0.020; the nitrogen appears to have captured about 2.8 times its "fair share." Although no realistic theoretical model has yet been developed to calculate this ratio, some European workers have come up with an assumption and a recipe that do reasonably well for BN and other molecules composed of low-atomic-weight elements. The heart of the assumption is that muons are captured by an atom in proportion to the number of loosely bound electrons around the atom, a quantity that can be computed in a simple way from the ionicity of the B-N bond. We conclude with a case in which this concept has provided the first experimental evidence for the direction of the dipole in the diatomic molecule nitric oxide (NO). Our muon team measured the capture ratio $c(N)/c(O)$ for two gas targets, one consisting of 5 atmospheres of nitrogen (N_2) and 5 atmospheres of oxygen (O_2) and the other consisting of 10 atmospheres of NO. The capture ratios came out to be 0.834 and 0.959, respectively, and the ratio of the ratios was 0.870; that is, the nitrogen in the NO captured a small excess of muons. By our assumption, then, the loose electrons are slightly displaced toward the nitrogen end of the molecule. A detailed theoretical calculation of the molecular structure has led to the

same charge orientation.

Fission Comes to the Group

The reader who has followed our account this far is likely to have acquired the impression that the nuclear and radiochemistry group was becoming rather large and diverse, with a range of programs and interests beyond that usually associated with a single discipline. As seen through the eyes of the participants this transformation has been undramatic but noticeable in many ways. It has reflected changes both within the community of members—in interests and capabilities—and outside—in the spectrum of national programmatic needs and the Laboratory's response to them. The transformation, like most transformations on the science and technology scene, had an important organizational facet: the question of how people and resources were to be organized and allocated for the strongest contribution to the Laboratory's goals. Consideration of the overall nuclear and radiochemistry effort from this standpoint received a great deal of attention and resulted, in late 1980, in the first major structural change since the group was formed in 1947. The nuclear and radiochemistry group, which had reached a membership of 100, was split into three groups, the first under Group Leader H. A. O'Brien and centered on research in medical radioisotopes, the second under Group Leader E. A. Bryant and centered on isotope geochemistry, and the third under Group Leader J. E. Sattizahn and centered on weapons diagnostics and other topics in nuclear and radiochemistry. In practice and as intended, the boundary lines are fuzzy. The groups continue to share the same building complex and facilities. Most important, they continue to enjoy the contacts and cross-fertilization of ideas that have contributed so significantly to past developments. ∎

Further Reading

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Jere D. Knight joined the Manhattan Project at the University of Chicago in 1942 after receiving a Bachelor of Science in chemistry from St. John's University (at Collegeville, Minnesota) and completing two years of graduate work at the University of Minnesota. In 1943 he transferred with the nuclear chemistry group under C. D. Coryell to Oak Ridge, where he worked on fission products, reactor neutron activations, and the first macro production of tritium. Following a return to the University of Minnesota and a Ph.D. in physical chemistry in 1948, he spent the next year and a half as an instructor in chemistry and research associate in physics at the University of Illinois. In 1949 he joined the nuclear and radiochemistry group at Los Alamos, where he has been involved in many of the developments described in this article. He was appointed a Laboratory Fellow in 1981. Since his retirement in 1982 he has served as a consultant to his former group.
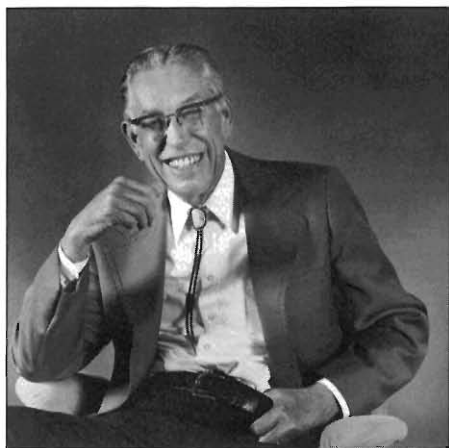
James E. Sattizahn received his B.S. in chemistry from Lawrence College in Appleton, Wisconsin in 1942 and joined the Manhattan Project at Oak Ridge in March 1944. (He and the co-author of this article worked only 100 yards apart at Oak Ridge, but they never met until both had moved to Los Alamos.) The Bikini tests brought Sattizahn to Los Alamos on June 5, 1946 as part of the team assembled to carry out radiochemical diagnostics. He was an original member of the Laboratory's permanent nuclear and radiochemistry group formed in early 1947 and has served as its Leader since January 1, 1971. He received a Ph.D. in radiochemistry from the University of New Mexico in 1957 under the Laboratory's graduate study program.